

SOURCE CONTRIBUTION OF LEAD IN HOUSE DUST FROM A LEAD MINING WASTE SUPERFUND SITE

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The relative contribution to household dust of lead particles from a mining waste superfund site and lead-based paint is investigated. Automated individual particle analysis (IPA) based on scanning electron microscopy (SEM) and X-ray energy spectroscopy (EDX) is used to develop a classification algorithm for determining lead particle source contribution in household dust vacuum bags. On a volume basis the proportion derived from the mining waste is found to be 26%, the proportion derived from a paint source is 16%, and the proportion from soil is 37%. In 15% of the lead particles identified a specific originating source could not be determined. Using a weighting method accounting for the lead concentration per particle rather than volume the contributions were similar for mining waste and paint, 21% and 23%, respectively, but the soil contribution was reduced to 8%, and the source for 29% of the lead could not be identified. These results suggest that the contribution of waste piles to the lead present in household dust is at least as important a source as paint. There is evidence to suggest that a large percentage of lead in the soil also originated from the waste piles and the overall contribution, therefore, of the waste piles may be greater than the contribution from paint.

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2. Abbreviations: ATSDR, Agency for Toxic Substances Disease Registry; cm², square centimeter; EDX, X-ray energy spectroscopy; IPA, individual particle analysis; ft², square feet; mg, milligram; ppm, part per million; Pb, lead SEM, scanning electron microscopy; µg, microgram.

3. Key words: exposure, household, lead, mining-waste, source-apportionment.

BACKGROUND

As part of a study of the Big River Mine Tailings Superfund Site Lead Exposure Study (ATSDR, 1997), Individual Particle Analysis techniques were applied to household vacuum cleaner bag dust from residential units and to the potential source materials, mine tailing waste and paint. Lead is naturally occurring in the area, but the deposition of mine tailings at ground surface has made lead exposure to people more prominent through use as fill material and wind blown deposition. Lead is also a health hazard in older homes where lead-based paint had been used. People living near the site have been exposed to lead through incidental ingestion of soils and dust contaminated with lead (ATSDR, 1994) and increased percentage of elevated blood leads in children when compared to a control site has been attributed to the presence of the mine tailings (ATSDR, 1997). The contribution of the lead mining waste, paint and other sources is important to know for exposure and risk assessment evaluation. These findings can be used for development of intervention approaches for reducing the exposure hazards, and setting priorities for prevention. The study objectives of this component were to: (1) determine whether lead particulate in the mining waste materials could be distinguished from other sources of lead-bearing origin; if so, (2) to develop a classification scheme that will distinguish between lead particulate originating from mining waste from that of paint; and, (3) to estimate the percent contributions of mine waste and paint to the lead present in household dusts. Presented are a summary of the findings.

Lead in household dust is derived from a variety of sources. The major potential source in residential properties is lead-based paint (Lanphear et al., 1996). Further contributions to dust lead loading include hobbies (soldering, ammunition reloading, etc.), lead derived from the work place environment, infiltration of atmospheric aerosols and transport (windblown, foot traffic, etc.) of contaminated soils into the home (Piacitelli et al., 1995; NIOSH, 1995). Given the variety of sources, it is reasonable to assume that particulate lead in household dust may exhibit a great diversity in its physical and chemical forms; aspects which bear on the uptake of lead in mammalian systems (Barltrop and Meek, 1979; Davis et al., 1992; Freeman et al., 1992; Dieter et al., 1993). Therefore, exposure studies are enhanced through estimates of lead source attribution. Inability to assess source contributions limits the reliability of exposure determinations. From a public health perspective this complicates decisions on intervention measures.

Source apportionment of lead in household dust, soil, and airborne particles from potentially contributing sources is difficult. In principle, the receptor model approach used for atmospheric aerosols (Friedlander, 1973; Dzuhay et al., 1984) could be applied, but environmental transformations of lead (Olson and Skogerboe, 1975; Johnson and Hunt, 1995) make this difficult for soils because the constant source composition assumption needed (Watson, 1982) is violated. Bulk chemical analysis of soils and dusts (Davies et al., 1985; Fergusson and Schroeder, 1985; Culbard et al., 1988) have lacked resolution, indicating merely that soil does contribute to house dust. The "best tracer method" of Stanek and Calabrese (1995) shows potential for quantifying the amount of soil lead in house dust, but has not been applied to such determinations. Similarly, stable isotope tracer methods (Yaffe et al., 1983; Robinowitz, 1987) have only had limited success. Radio isotope studies are prone to potential problems of source blurring due to mixing of lead from various sources.

A third method, automated individual particle analysis (IPA), was selected for source apportionment in the present study. IPA based on scanning electron microscopy (SEM) and X-ray energy spectroscopy (EDX), has been used to assess the potential originating sources of the lead. These techniques have been shown to discriminate between lead particles at the individual level when bulk sample analysis indicate compositionally similar products (Hunt et al., 1992; Vander Wood, 1993). Chemical and elemental morphology and composition is determined through SEM and EDX analysis. Particles with morphologies and elemental associations' characteristic of different particulate lead source types can be identified and enumerated. If a classification scheme for IPA results can be developed that provides distinctive "signatures" for the different source type materials, it can be applied to other related samples analyzed under identical conditions, providing a descriptive source apportionment (Hunt et al., 1991).

Particulate lead transported to, or originated within, an interior residential environment does not appear, subsequently, to undergo significant chemical transformations that alter its "original" physicochemical form. Once a particulate has entered the indoor environment it is protected from geochemical weathering. Based on knowledge of product composition and potential degradation products, groups of particles that most likely are derived from the same source can be probabilistically identified on the basis of morphology and composition. This automated microscopy-based method has been used in the United Kingdom as part of a comprehensive study of lead contamination in environmental dusts (Thorton et al., 1994), for the source apportionment of lead in house dusts (Hunt et al., 1994), as part of a lead contamination study in Port Pirie, Australia (Body et al., 1988) and in studies to determine lead sources bear a lead smelter in Missouri (Vander Wood and Brown, 1992). At present, this method generates essentially semi-quantitative results, but should be sufficient for discriminating between lead derived from paint alone or other environmental sources, such as mining waste piles (Johnson and Hunt, 1995).

METHODOLOGY

Field Samples Characterized For Classification Scheme

Composites from five mining waste piles, randomly selected lead-based paint chip samples collected from study area homes, and eight yard soil composites were characterized and used in development of the classification scheme to determine particulate lead origin in household dust. The results of the classification scheme was applied to household dust samples: eight homes selected randomly from 25 homes from the study area, and two homes from control areas. The household vacuum bags were collected from the occupant vacuum cleaners during previous environmental sampling (ATSDR, 1997). Eligibility was determined by the presence of lead in each of the following components at elevated levels: (1) the household average of lead-based paint on friction surfaces (such as windows and doors) within a child's bedroom and primary indoor play areas was $\geq 1.0 \text{ mg/cm}^2$; (2) samples of yard soil composite more than five feet from painted outside walls which contained $\geq 400 \text{ ppm}$; (3) composite sample of household vacuum cleaner bag dust $\geq 200 \text{ ppm}$; and, (4) interior window sill wipe samples $\geq 500 \text{ } \mu\text{g/ft}^2$.

IPA Characterization

The individual particle analysis protocols detailed below for specimen preparation and scanning electron microscope characterization were derived from those of Hunt et al. (1992) and Johnson and Hunt (1995). Since, in general, only a few percent of the particles in soils or house dusts contain detectable lead by x-ray microanalysis, the analytical challenge for such microscopy-based approaches is to provide a time efficient characterization of statistically significant populations of features. Our analytical strategy employed size fractionation of the environmental samples to optimize instrumental conditions; more rapid and accurate feature location is possible for a limited size range. It also utilized a particle search strategy of "high" thresholding that selects high average atomic number features for characterization, usually excluding from analysis such geologic materials as quartz, clays or limestone. Lead containing features aggregated with such particles or present on them in the form of a precipitation rind would, however, be identified and analyzed by this procedure.

Paint samples, randomly selected from study homes, and one specimen from a Syracuse house, were ground with a micro mortar and pestle and deposited directly on polycarbonate membranes affixed to graphite SEM stubs. Composite samples from waste pile (Chat) collections and soil collections were sieved through an 85 μm opening monofilament polyester mesh. After homogenization, sub-samples of this material were sonicated in aqueous solution and portions were filtered onto 0.4 μm pore size polycarbonate membranes and mounted in a similar fashion. Composites of vacuum cleaner bag contents from the homes were subjected to wet sieving (10% ethanol solution) through the 85 μm mesh during application of ultra sound to separate inorganic particles from the fibrous matrix. Portions of the separated material suspension were filtered through polycarbonate membranes for analysis. For all SEM preparations, mass loading was adjusted to obtain a monolayer of particles with sufficient space between features to minimize adjacent particle X-ray fluorescence. All specimens were coated with carbon in a high vacuum evaporator prior to analysis.

Each feature characterized by the IPA procedures contains information on size, estimated volume, and the relative X-ray emission intensity for 25 different elements. The X-ray spectrometer regions of interest and the net count corrections, peak overlap corrections and elemental efficiency factors were developed using NIST SRM 1633 (fly ash), USGS Standard BCR-I, and NIST SRM 2710 (metals in soil). Using a procedure developed by Johnson et al. (1981), the estimated volume of each feature, an assigned specific gravity, the relative X-ray intensity of each particle and the expected molecular form of occurrence of each element for a given particle type were summed over all observations to compute a bulk chemical composition. The spectrometer set up we used gave acceptable results for the major element matrix of SRM 2710 (Johnson and Hunt, 1995) and the lead concentration so determined was within 10% of the certified value. The percentage of net X-ray counts contributed by each element monitored in each feature was used for developing the particle classification criteria.

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The instrumentation employed for the IPA characterizations was an ETEC AutoScan SEM interfaced with a KEVEX 7500 X-ray spectrometer system and controlled by a LeMont Scientific DA-IO Image Analysis System. All analyses were carried out at a magnification of 300 \times and a digital scan generator pixel density sufficient to characterize particles as small as 0.5 μm . A few micro-crystals of NaCl were deposited on the edge of each specimen as an imaging standard. The backscatter electron signal imaging threshold was adjusted so that sodium chloride just disappeared. Thus the features characterized were limited to those with average atomic number 14 and higher. Limited normal thresholding analyses of the waste pile material were also carried out to provide a general description of the particle types they contained.

RESULTS AND DISCUSSION

Analytical Limitations

The analytical results and their interpretation relative to the potential for children's exposure to lead in the study homes are of course limited by the context of the analytical techniques. We emphasize that our observations are limited to particulate phases smaller than 85 μm in size. However, inadvertent ingestion of soils and dusts through the normal hand-to-mouth activity of children is a major exposure pathway, and the size of particles on children's hands is generally smaller than 100 μm (Duggin et al., 1985; Hunt, 1994; Wang et al., 1994). Characterization of the size range distribution of household dust captured by occupants vacuum cleaner bags in an urban area showed 50% of the total dust and lead containing dust mass was in the size fraction less than 63 μm (Sterling, 1998). Other investigators have found similar results. Que Hee et al., (1985) reported that 77% of the lead in total household dust samples was less than 149 μm , and 90% of the particles which adhere to a child's hands less than 10 μm . The log normal geometric mean (lnGM) area equivalent diameter for the lead bearing features found in "high" thresholding of chat and soil samples were 3.0 μm and 1.6 μm , respectively. Individual home dust samples lnGM ranged from 1.2 μm to 2.9 μm with a combined lnGM of 2.02 μm , and all showed log normal distributions. Thus, characterization of the sub-85 μm fraction would seem to carry a substantial proportion of the information relative to potential exposure sources.

"High" thresholding for particle location offers a substantial advantage for time efficiency in the particle analysis. For instance, under normal thresholding conditions where all inorganic particles are characterized, lead associated with the waste pile materials was observed in less than 6% of the features. Under "high" thresholding, 60–80% of the particles located contained detectable lead. Analysis of several hundred lead-containing features could be accomplished in about 2 hours as opposed to 20+ hours using normal thresholding. For the house dusts, where bulk lead concentrations were significantly lower than those for the waste pile materials, this time saving was necessary for the analysis to be feasible.

The accuracy of locating lead-bearing features is also increased by "high" thresholding. The software controlling the SEM beam directs it to the geometric center of a feature image, and gathers X-ray information from a circle whose diameter is one half the distance from the geometric center of the image to the nearest edge. Lead-bearing inclusions, rinds or small lead rich particles aggregated to larger features might not be analyzed with this strategy under normal thresholding

conditions; they would be located by "high" thresholding. Only the high atomic number portions of a heterogeneous particle would be characterized in the latter case and the size measured for a lead-bearing feature would not accurately reflect the size of the entire particle. Application of ultra sound to disaggregate features during specimen preparation might also affect the size distribution of some particle agglomerations. In the case of the vacuum cleaner samples, however, sonication seemed the most accurate way of separating the inorganic particles from the organic matrix.

Particle Classification Scheme Development

The waste pile materials and the paint samples showed distinct particle types. Under conditions of both "normal" and "high thresholding" analysis the percentage of lead X-rays in the paint particles showed a similar distribution and generally contained titanium, barium and/or zinc as distinctive co-constituent elements. Various amounts of calcium, sulfur and silicon were also found. An initial characterization of the waste pile material, at normal thresholding, showed that on a volume weighted basis it contained: 76% calcium rich particles (as in limestone and dolomite), 11% calcium/silica mix particles, 9% high silica, 1% as high iron particles (including pyrite) and about 3% miscellaneous features. Only trace amounts of aluminosilicate materials were observed. Lead was observed in less than 6% of the features. Under high thresholding, lead was observed in 60-80% of the waste pile particles and had a strong association with calcium, and to a lesser extent with particles containing iron and sulfur. In general, lead X-ray relative intensity in these features were either low (less than or equal to 15%) or high (greater than 50%).

A descriptive, hierarchical sorting algorithm for particle classification was developed based on these generalities. Figure 1 shows the structure of the scheme and indicates the specific X-ray intensity criteria for the elements found useful in classification. Six major categories are delineated on the left side of the figure. A particle satisfying one of these categories was further partitioned by the second set of sub-category criteria outlined to the right. No iron-sulfur-lead (Fe-S-Pb) particle elemental associations were observed in the paint samples for category 1, so no further sub-classification was developed, and category 1 is only characteristic of waste pile materials. Analyses of the paints and the waste pile materials were used, in an iterative fashion, to establish waste pile and paint sub-categories for categories 2-6 minimizing the degree of incorrect classification. That is, criteria were sought which excluded Chat particles from the Paint subgroups and vice versa. This emphasis on preventing incorrect classification is the chief value of a descriptive classification scheme. However, it creates an "unresolved" class for each major group; the unresolved category contains observations common to both types of source material.

Table I shows results of applying the sorting scheme to the potential source materials. The rightmost column indicates the number of lead-bearing particles characterized for each specimen. The data are presented two ways: (1) as percentages of the total volume of lead-bearing features characterized in each sample (which will be referred to as "volume-weighted") and are derived from the sum of the classification results (Figure 1); and, (2) computed in similar fashion, but normalized to the sum of feature volume times lead X-ray relative intensity percentage (which will be referred to as "concentration-weighted"). The former corrects for the different estimated volumes of lead bearing features. The latter adjusts for estimated lead content as well as volume and is considered a more useful estimate of exposure potential.

Category

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FIGURE
potential
Values

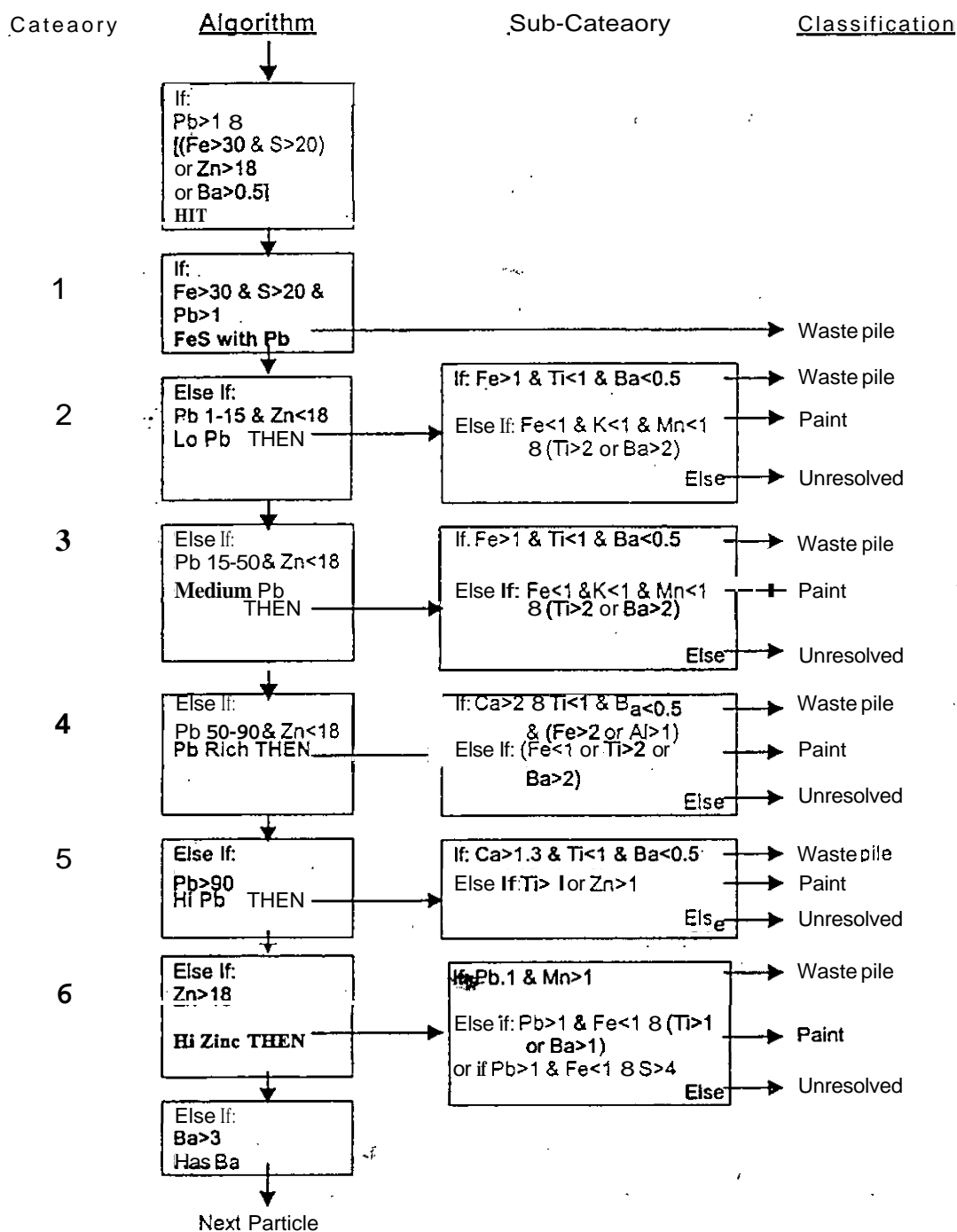


FIGURE 1. Criteria used in linear sorting algorithm for the classification of particles identified in the potential source materials and household dust samples using the "High" thresholding search technique. Values represent percentages of the net X-ray count obtained for each particle.

Chat material sample results under the paint column of Table 1 indicate that on a volume-weighted basis less than 5%, and on a concentration-weighted basis less than 7%, of the waste pile lead is incorrectly classified as being of paint in origin. The paint sample results under the waste pile column show that generally less than 5% of the lead is improperly ascribed for both weighting methods. The unresolved component in each of these materials is quite variable. While results exhibit considerable variability in the "signatures" of the possible contributing source materials, the overall median percent identification for volume and concentration-weighted as the actual source is high for both waste pile (69% and 79%, respectively) and paint (82% and 86%, respectively) samples, and are similar for both weighting methods. To account for possible limitations in sample size and individual extreme values the medians are reported to describe the central tendency of the distributions.

TABLE 1. Particle Classification Results Given by the Sorting Algorithm (Version 1.0) for Source Materials. Results Are Normalized First as a Percentage of the Total Estimated Volume of Lead-bearing Features Found in Each Specimen (Percent-volume), and Second as a Percentage of Total Estimated Feature Volume Weighted by the Lead X-ray Relative Intensity for Each Particle (Percent-concentration)

	Waste Piles		Paint		Unresolved		Number of particles ¹
	Percent Volume	Percent Concentration	Percent Volume	Percent Concentration	Percent Volume	Percent Concentration	
Chat							
glass	47.8	22.0	3.8	5.1	48.3	72.9	282
lw385b	79.1	62.4	4.6	6.8	16.3	30.8	210
brc101	74.1	69.4	3A	3.8	22.4	26.8	450
lwffc	95.6	73.5	0.5	4.7	3.9	21.8	307
brffc	90.0	86.2	0.2	0.2	9.8	13.6	302
Soil ²							
soil1	58.5	50.5	0.0	0.0	41.5	49.5	178
Paint							
P431ia	0.0	0.0	92.2	96.0	7.8	4.0	272
P256ia	0.1	0.1	25.0	36.3	74.9	63.6	272
P511p4	0.1	0.1	99.1	98.9	0.8	1.0	280
P480p1	0.1	0.1	93.4	89.4	6.5	10.5	469
P509p2	0.5	0.3	69.3	65.7	30.3	34.0	386
P495p2	0.2	0.3	93.0	88.4	6.7	11.3	575
djp001	0.2	0.3	91.2	92.7	8.6	7.0	395
stp003	0.6	0.6	75.2	82.9	24.2	16.4	389
P256xa	1.6	1.7	9.7	12.8	88.7	85.5	279
P172xa	2.0	2.2	74.9	77.9	23.1	20.5	290
P172xb	2.6	2.5	42.4	42.3	55.2	55.2	287
P506p1	5.3	5.3	89.3	88.9	5.4	5.8	400

¹Number of particles that lead-bearing features were found for 'high' thresholding.

²Yard soil samples from the eight homes were pooled together to form one sample for P A analysis.

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The soil data show no particles which could easily be identified as having been derived from paint, but about half of the lead volume and concentration-weighted particles may be of waste pile material origin (Table 1). Since paint sources were not expected in the soil samples this suggests that the algorithm is not prone to false positive indications for paint. Additionally, previous investigations have found elevated soil lead concentrations above background and concluded the waste piles were the primary source (ATSDR, 1994, 1997). For the waste pile materials, paints P172xb, P256ia and P256xa, and for the composite soil sample, most of the unresolved entries in Table 1 were contributed by class category 5 particles (Figure 1) in which the lead X-rays were greater than 90% of the total net count. Such features could be lead metal, various lead oxides or lead carbonate particles; particles that may be naturally present in paint compositions, may form in the waste pile materials and soil as the result of environmental transformations, or may be derived from other sources, and be transported indoors. Without additional instrumental capabilities, we could not distinguish among these possibilities and they reflect a limitation of the present study.

Hunt et al. (1991) used a similar iterative procedure for development of a descriptive classification scheme applied to lead in house dust samples. In their work, automotive lead categories were included where the presence of bromine and lead in the X-ray microanalysis of particles indicated lead halides characteristic of automobile emissions. We did not include such classes as we did not observe any such characteristic features in the samples we analyzed. It is possible that unresolved particles of both the soil and the house dust samples contain lead of automotive origin.

Source Apportionment Model

Application of the initial classification scheme to the house dusts derived from home vacuum cleaners is shown in Table 2. The descriptive classification for concentration-weighted results showed significant contributions to the ambient house dusts by both the waste pile materials (1–34%, median 17%) and the lead based paints (1–50%, median 12%), but over 60% of the lead (32–99%) was unresolved. Repeat analysis on a second prepared sample from house dust vacuum cleaner bag H465 indicated good precision of the method for application to the field samples from the superfund area, particularly for the concentration-weighted results which varied by less than 10%. The classification scheme was also applied to two samples not derived from the superfund area: sample H314 a control home matched for demographics (ATSDR, 1997), and SYR collected separately as a preabatement sample from a residence in Syracuse, NY. Results from both samples showed limited indication of contribution from waste pile sources or paint, with the majority of classification results as unresolved.

The data set for source particle characterization is not yet large enough to support a classification scheme with substantially greater resolution, but the results can be used in a simple source apportionment calculation. Since all of the potential source materials contain unresolved constituents, such a model might be capable of attributing portions of the unresolved components to characteristic sources.

TABLE 2. Particle Classification Results Given by the **Sorting Algorithm** (Version 1.0) for Home Vacuum Cleaner Dusts. Results are **Normalized First** as a Percentage of the Total Estimated Volume of Lead-bearing **Features** Found in Each Specimen percent-volume), and **Second** as a Percentage of Total Estimated Feature Volume Weighted by the Lead X-ray Relative Intensity for Each Particle (Percent-concentration)

	Waste Piles		Paint		Unresolved		Number of particles ¹
	Percent Volume	Percent Concentration	Percent Volume	Percent Concentration	Percent Volume	Percent Concentration	
H465	40.6	21.0	16.1	24.4	43.3	56.5	279
H465rep ²	27.1	21.9	24.2	25.6	48.7	52.5	323
H256	15.5	15.0	24.2	29.2	60.3	55.8	171
H431	55.7	31.8	0A	0.6	43.9	61.6	177
H172	47.7	14.7	10.8	11.7	41.5	73.6	235
H275	50.2	17.3	27.9	50.3	21.9	32.4	168
H404	48.8	25.8	17.2	41.7	34.0	32.4	146
H474	27.0	14.2	0.5	0.9	12.5	84.9	156
H282	46.8	33.8	2.8	3.6	50.4	62.6	163
H314control ³	11.2	0.8	6.5	2.4	8.23	98.8	10
SYRcontrol ⁴	6.4	4.3	5.8	4.3	8.23	85.6	302

¹Number of particles that lead-bearing features were found for 'high' thresholding.

²Repeat sample analysis

³Control home remote from the superfund study Site. Bulk analyses showed concentrations < 200 µg/Kg of lead, and only 10 lead-bearing features were found after analysis of 665 high average atomic number features.

⁴Study residence in Syracuse, NY sampled prior to lead paint abatement activities.

Source apportionment models based on **chemical mass balance** can be developed for any data partitioning approach; elemental composition, quantification of crystalline components, isotope ratio signatures, etc. (Watson, 1982). Johnson and McIntyre (1982) showed that the mathematical methods could be used for a particle class balance. After developing a **sorting** scheme for classifying individual particle analysis results, it was applied uniformly to both potential source particles and ambient aerosol particles. From this, characteristic particle type distributions for potential source materials established a source signature matrix analogous to that from bulk elemental composition determination. The general form of the mathematical relationship is:

$$C = A * S \quad (1)$$

where C is a column matrix of particle types in the unknown or ambient sample, A is a column matrix of the (potential) source signatures, and S is a column matrix of the source strengths. A short FORTRAN routine was used to solve for S:

$$S = (A * A^T)^{-1} * A^T * C \quad (2)$$

This least squares procedure computed the fraction of each analyzed source necessary to produce the ambient sample signature, minimizing the difference between observed and predicted particle type distributions.

The particle class balance model for apportionment is subject to the same assumptions as the chemical mass balance; (1) the number of components measured is greater than the number of sources to be fit, (2) the source composition completely describes the ambient samples, (3) the source compositions are constant, and (4) the sources are linearly independent of each other. Without a complete characterization of each point or area source for each study home, we can't address the constant source composition assumption. We used average signatures for the three types of source materials sampled (chat, soil, paint). The analytical results for each group were combined and classified according to the scheme in Figure 1 to provide the source signature matrix. The source independence is also subject to question. Particulate lead deposited in soils undergoes a variety of transformations altering its (possibly) characteristic form as emitted from a source. Until further investigation of the unresolved category 5 type particles, we have chosen to include an additional generic source for these particles in the particle class balance source signature matrix calling it "common oxide" with a classification entry of 100% in category 5.

Particles with undetermined source were found in significant amounts in many of the source material characterizations (Table 1) and were dominant in the analytical results of the household dust samples. Ascribing their origin in the latter samples to a probable source was carried out by a least squares apportionment model. Average particle classification signatures (distribution across the 16 classes in the sorting algorithm) were computed for waste pile materials, paints, and the composite soil results. A fourth signature for the common (unresolved) oxide type particles was also included in the source signature matrix; and apportionment was carried out using both the volume-weighted and concentration-weighted summaries.

Applying the least squares apportionment model to the household vacuum dust, shown in Figure 2, demonstrate how the particulate lead in the collected house dust samples are distributed across four possible source types. Negative entries arise from the least squares fitting procedure, and for the results presented here, are less than 10%, and indicate that the uncertainty associated with the model results is low. Based on volume weighted signatures, waste pile sources show greater medium contributions than paint particles, 36% with a range of -8% to 57% and 16% with a range of 2% to 80%, respectively. Transport of soils contributed about 36% (1% to 63%), and the common oxide was 15% (-3% to 38%). By comparison, the 15 house study of Hunt et al. (1994) found that on a volume weighted bases (< 64 μ m fraction) paint contributed about 33% (5% to 95%) to the house dust and the common oxide particle type about 4% (0 to 14%). In the latter study, soils and mad dusts together, as external sources, accounted for 46% (1% to 80%) of the particulate lead; in the present work, soils and waste pile materials together average 62% in their contribution. In both studies, the source contributions were highly variable across potential types.

Apportionment results based on the concentration-weighted summaries for waste pile material and paint contributions are similar to those of volume weighting alone (Figure 2), showing a median of 21% (6% to 60%) for waste pile particles and 23% (-1% to 89%) for paint. However, the soil contribution is substantially less, about 8% (-2% to 19%) and the common oxide source strength is higher at 29% (4% to 75%). These differences are the expected results since the common oxide features have a high lead content and much of the lead in soils is bound to larger particles (Johnson and Hunt, 1995).

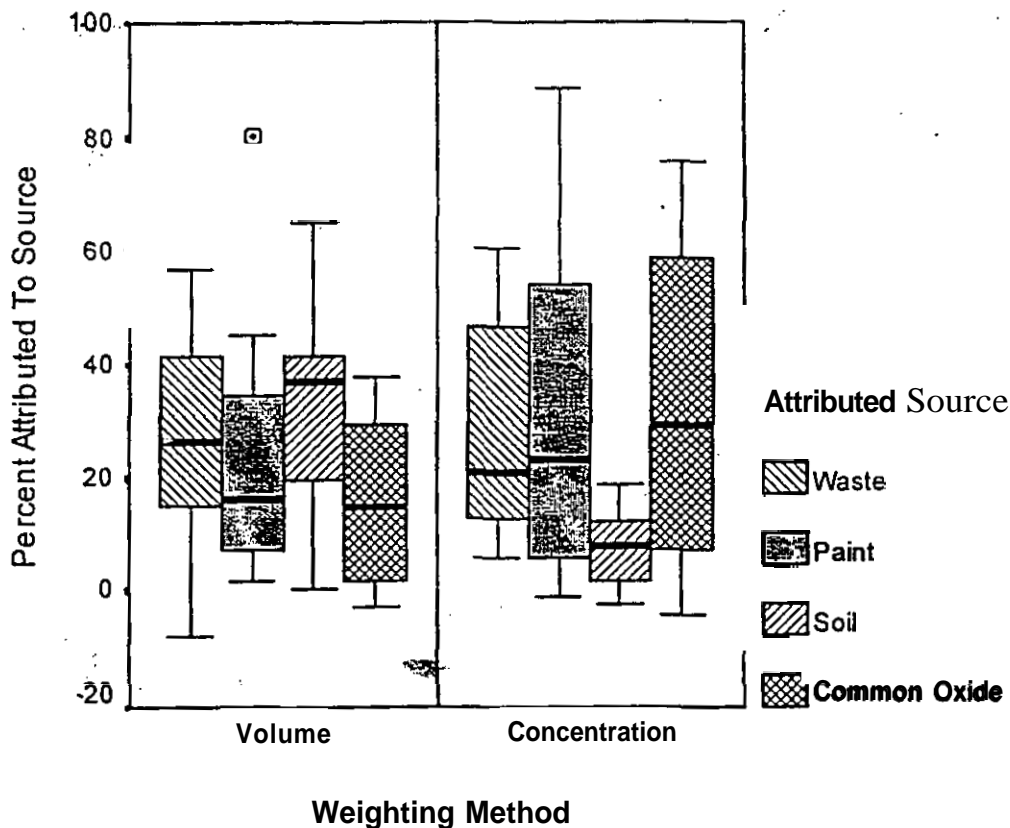


FIGURE 2. Boxplot describing results of applying least squares results of the classification/sorting algorithm to the interior household dust samples. Results are first normalized and weighted as a percentage of the estimated volume of lead-bearing features found in the attributed source, and second weighted as a percentage of total estimated concentration. The box contains 50% of the data. The crossbar within the box indicates the median. Whiskers above or below the box describe the range and skewness for 95% of the data distribution. The circles show individual outliers beyond the 95th percentile.

CONCLUSIONS

Using the IPA method and applying a least squares apportionment model, an analysis of lead sources on the household vacuum bag dust at a mining waste superfund site was performed. Based on particle volume-weighting the median proportions of lead derived from the contributing sources is 26% from mining waste, 16% derived from a paint source, and 37% from soil. For 15% of the lead particles a specific originating Source could not be determined. Using the particle concentration-weighting method the median proportions observed were 21% from mining waste, 23% from paint, 8% from soil, and 29% could not be identified. These results suggest that the waste piles are at least as important a contribution source as lead-based paint to the presence of indoor lead dust. It is reasonable to assume that a large percent of the lead derived from yard soil and the unresolved or common category also originated from the waste piles. Therefore, the overall contribution of the waste piles to house dust may be greater than the contribution from paint, by both total particle volume and lead concentration-weighted methods. Knowledge of the proportion of exposure from contributing sources is important in assessing exposure, health risk, and development of health promotion activities.

To add additional resolution in the descriptive classification scheme and to more fully determine the impact on lead in household dust derived from the waste pile and the contribution of soil as a source and/or transport media, further characterizations need to be performed on particle morphology (volume and size) and particle concentration of lead found in household dust.

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REFERENCES

- AGENCY FOR TOXIC SUBSTANCES AND DISEASE REGISTRY (ATSDR) (1994). *Preliminary Public Health Assessment for Big River Mine Tailings Site, Desloge St. Francois County Missouri*. Agency for Toxic Substances and Disease Registry, U.S. Department of Health and Human Services, Atlanta, GA.
- AGENCY FOR TOXIC SUBSTANCES AND DISEASE REGISTRY (ATSDR) (1997). *Big River Mine Tailings Superfund Site Lead Exposure Study: Report to the Agency for Toxic Substances and Disease Registry, U.S. Department of Health and Human Services, Atlanta, GA.*
- BARLTROP, D. and MEEK, F. (1979). "Effect of particle size on lead absorption from the gut." *Arch. Environ. Health* July-August, 280-285.
- BODY, P.E., INGILS, G.R., and MULCHAY, D.E. (1988). "Lead contamination in Port Pirie, South Australia." *South Australia DEP Report Number 101*, April 1988.
- CULBARD, E.B., THORNTON, L., WATT, M., WHEATLEY, S., MOORECROFT, S., and THOMPSON, M. (1988). "Metal contamination in British urban dusts and soils." *J. Environ. Quality* 17(2):226-236.
- DAVIS, A., DREXLER, J.W., RUBY, M.V., and NICHOLSON, A. (1993). "Micromineralogy of mine wastes in relation to lead bioavailability, Butte, Montana." *Environ. Science Technol.* 27(7):1415-1425.

- DIETER, M.P., MATTHEW, H.B., JEFFCOAT, R.A., and MOSEMAN, R.F. (1993). "Comparison of lead bioavailability in F344 rats fed lead acetate, lead oxide, lead sulfide or lead ore concentrate from Skagway, Alaska." *J. Toxicol. Environ. Health* 39:79-93.
- DZUBAY, T.G., et al. (1984). "Interlaboratory comparison of receptor model results for Houston aerosol." *Amos. Environ. 18*(8):1555-1566.
- FREEMAN, G.B., JOHNSON, J.D., KILLINGER, J.M., LIAO, S.C., FEDER, P.I., et al. (1992). "Relative bioavailability of lead from mining waste soil in rats." *Fund. Appl. Toxicol.* 19:388-398.
- FRIEDLANDER, S.K. (1973). "Chemical element balances and identification of air pollution sources." *Environ. Science Technol.* 7:235.
- FERGUSON, J.E. and SCHROEDER, R.J. (1985). "Lead in house dust of Christchurch, New Zealand: Sampling, levels and sources." *Science Total Environ.* 46:61-75.
- HUNT, A., JOHNSON, D.L., and THORNTON, I. (1991). "Descriptive apportionment of lead in house dust by automated SEM." *Water, Air, Soil Pollut.* 57-58:69-77.
- HUNT, A., JOHNSON, D.L., WATT, J.M., and THORNTON, I. (1992). "Characterizing the sources of particulate lead in house dust by automated scanning electron microscopy." *Environ. Science Technol.* 26(8):1513-1522.
- HUNT, A., JOHNSON, D.L., THORNTON, I., and WATT, J.M. (1994). "Apportioning the sources of lead in house dusts in the London borough of Richmond, England." *Science Total Environ.* 138:183-206.
- JOHNSON, D.L. and MCINTYRE (1982). "A particle class balance receptor model for aerosol apportionment in Syracuse, NY." In: *Proceedings, Receptor Models Applied to Contemporary Pollution Problems*. Air Pollut. Control Assoc. Specialty Conference SP-48, pp. 238.
- JOHNSON, D.L. and HUNT, A. (1995). "Analysis of lead in urban soils by computer assisted SEM/EDX—Method development and early results." In: *Lead in Point, Soil and Dirt: Health Risk, Exposure Studies, Control Measures, Measurement Methods, and Quality Assurance*. (M. Beard and E. Allen Iske, eds.). ASTM STP 1226.
- LANPHEAR, B.P., WEITZMAN, M., WINTER, N.L., EBERLY, S., YAKIR, B., TANNER, M., EMOND, M., and MATTE, T.D. (1996). "Lead contaminated house dust and urban children's blood lead levels." *Am. J. Public Health* 86:1416-1421.
- NATIONAL INSTITUTE OF OCCUPATIONAL SAFETY AND HEALTH (NIOSH) (1995). "Construction workers exposed to lead are contaminating their homes and cars." *Asbestos and Lead Abatement Report*. June 587.
- OLSON, K.W. and SKOGERBOE, R.X. (1975). "Identification of soil lead compounds from automotive sources." *Environ. Science Technol.* 9:227-230.
- PIACITELLI, G., WHELAN, E., EWERS, L., and SIEBER, W. (1995). "Lead contamination in automobiles of lead-exposed bridgeworkers." *Am. Occup. Environ. Hyg. J.* 10(10):849-855.
- QUE HEE, S.S., PEACE, B., CLARK, S., BOYLE, JR., BORNSCHEIN, R.L., and HAMMOND, P.B. (1985). "Evolution of efficient methods to sample house dust and hand dust in the homes of children." *Environ. Res.* 38:77-95.
- RABINOWITZ, M. (1987). "Stable isotope mass spectrometry in childhood lead poisoning." *Biol. Trace Element Res.* 12:223-229.
- STERLING, D.A. (1998). Unpublished data: Saint Louis University School of Public Health, St. Louis, MO.
- STANEK, E.J., III and CALABRESE, E.J. (1995). "Soil ingestion estimates for use in site evaluation based on the best tracer method." *Human Ecol. Risk Assess.* 1(2):133-156.
- THORNTON, I., et al. (1994). "Lead contamination of UK dusts and soils and implications for childhood exposure: An overview of the work of the Environmental Geochemistry Research Group, Imperial College, London, England 1981-1992." *Environ. Geochem. Health* 16(3/4):113-122.
- VANDER WOOD, T.B. and BROWN, R.S. (1992). "The application of automated scanning electron microscopy/energy dispersion x-ray spectrometry to identification of lead rich particles in soil and dust." *Environ. Choices Tech. Suppl.* 1(1):26-32.
- VANDER WOOD, T.B. (1993). "Automated particle analysis in electron microscopy." *Proc. 51st Annual Meeting of the Microscopy Society of America, San Francisco, CA.* pp. 738-739.
- WANG E.Y., WILLIS, R.D., BUCKLEY, T.J., RHOADS, G.G., and LIOY, P.J. (1996). "The relationship between the lead concentration and the particle size of household dusts collected in Jersey City residences." *Appl. Occup. Environ. Hyg. J.* 11(3):199-205.

WATSON, J.
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SP-48,
YAFFE, Y.P.
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ZIMDAHL,

Manuscript
Manuscript

- WATSON, J.G. (1982). "Overview of receptor model principles." In: *Proceedings. Receptor Models Applied to Contemporary Pollution Problems*. Air Pollution Control Association, Specialty Conference SP-48, pp. 368.
- YAFFE, Y.P., FLESSEL, C.P., WESOLOSKI, J.J., et al. (1983). "Identification of lead sources in California children using the stable isotope ratio technique." *Arch. Environ. Health* 38:237-245.
- ZIMDAHL, R.L. (1977). "Behavior of lead in soil." *Environ. Science Technol.* 11:1202-1207.

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